HIPPO: A New High-Intensity Neutron Diffractometer for Characterizing Bulk Materials

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A new time-of-flight (TOF), high-pressure preferred orientation (HIPPO) neutron diffractometer for materials studies is under construction at LANSCE's Manuel Lujan Jr. Neutron Scattering Center. The majority of HIPPO was designed and manufactured at LANSCE and is part of a $42 million upgrade project of LANSCE facilities sponsored by the Department of Energy Offices of Basic Energy Science and Defense Programs. The development of the HIPPO instrument is a combined effort between the University of California campuses and national laboratories to attain scientific excellence in neutron diffraction, to advance our present knowledge of condensed matter, and to make neutron diffractometry an accessible and available tool to the national user community. HIPPO will allow researchers and students to conduct, for example, real-time structural studies in situ at high and low temperatures and at high pressures under a variety of environmental conditions. This unique neutron diffractometer will cover a broad variety of disciplines, including materials science and engineering, earth sciences, physics, and chemistry, and it will advance research in three interdependent material properties—high pressure, texture, and magnetism—all of which can be measured simultaneously with HIPPO. Between 150 and 250 experiments will be accommodated each year during an eight-month run cycle to benefit the neutron specialists and the materials- and earth-sciences communities. By providing easy and sustained access to HIPPO, we will open neutron diffraction to graduate-student thesis research, which often cannot wait until the formal proposal for a single experiment is approved. Use of the instrument will be free, and a user program is scheduled for summer 2001.

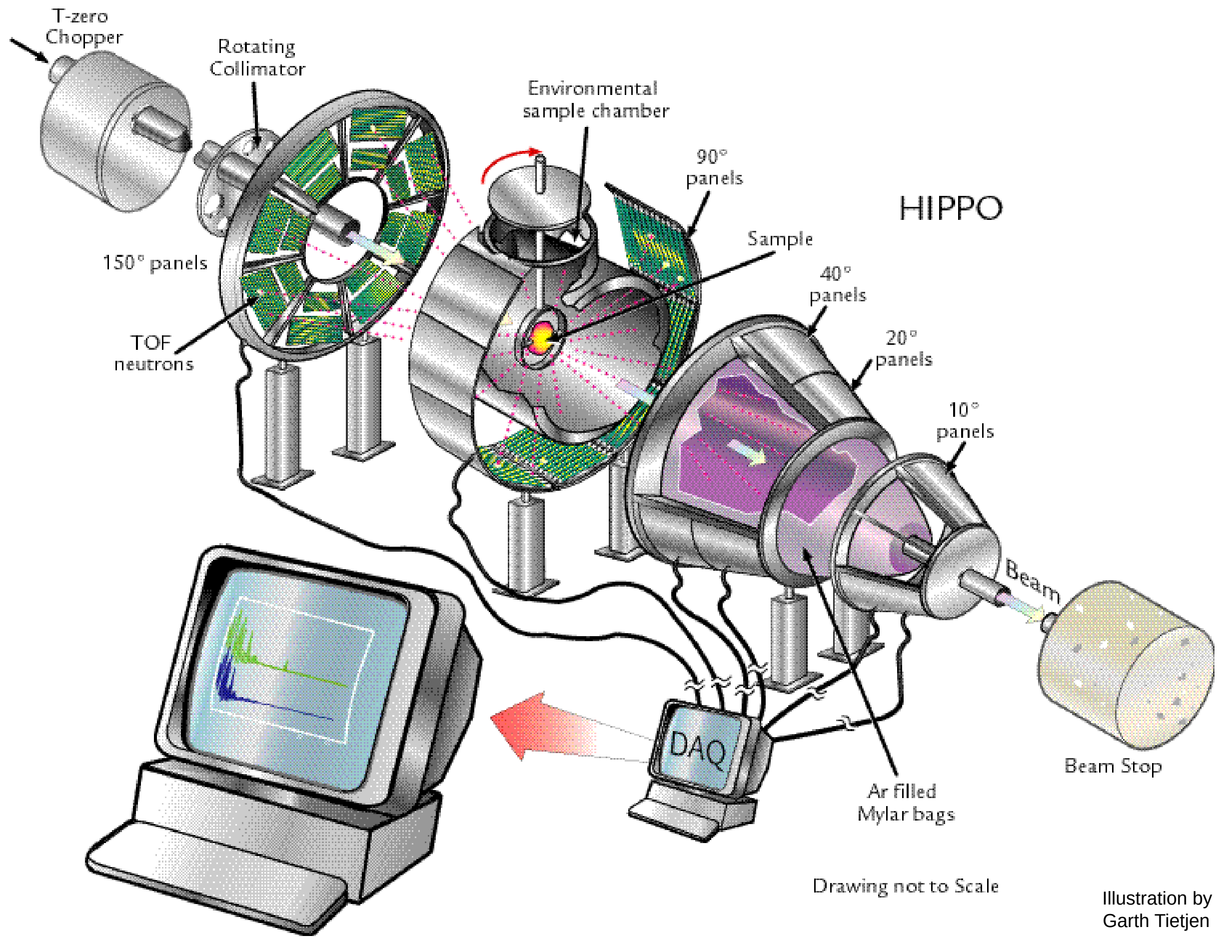
Advantages of Neutron-Diffraction Applications

Important new neutron-diffraction applications have emerged in both applied and fundamental condensed-matter research. Of particular interest to the research community is the investigation of small (1 mm^3) and large (2 cm^3) sample volumes at high (< 2,000 K) and low (> 10 K) temperatures, at high pressure (< 30 GPa), and in different atmospheres. Because neutrons can travel large distances through most materials without being absorbed, these tiny probes can be used to study atomic structures in bulk samples under a variety of experimental conditions. The availability of high spectral resolution also allows the simultaneous measurement of, for example, refined crystal structures, phase proportions, textures, and strains using modified Rietveld methods.[1,2]

The new high-pressure preferred orientation (HIPPO) diffractometer takes advantage of the improved neutron source at LANSCE and a short flight path (flux at the sample will be nominally 10^8 $n/cm^{-2}/s^{-1}$) to overcome a major limitation of neutrons—their weak intensity. The count rate for some experiments will be approximately 20 to 60 times what is currently obtained on the high-intensity powder diffractometer presently at the Lujan Center, and researchers will be able to make measurements in as little as 5 to 10 s. It will produce, within minutes rather than hours, time-of-flight (TOF) neutron-diffraction patterns from the data output of three-dimensional detector banks consisting of about 1,400 ^3He gas-filled detector tubes. HIPPO's fast data-acquisition capability and the novel arrangement of its detectors will allow researchers to study time-dependent processes and crystal orientations in bulk samples. With the analysis of the diffraction patterns using Rietveld codes, HIPPO will fast become the instrument of choice in the fields of phase transformation, high-pressure research, polycrystal anisotropy (texture-stress-strain), and complex materials crystallography. Researchers will be able to study the dynamics of reactions, recrystallization, and deformation of bulk anisotropic properties under various temperature and pressure conditions. No existing instrument has this capability.

State-of-the-Art Technology

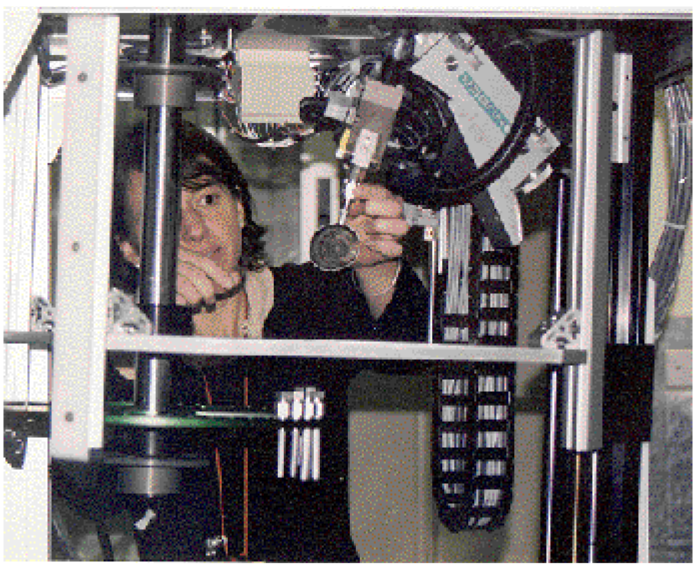
A state-of-the-art target-moderator system produces pulsed 'thermal' (slow) neutrons that travel down HIPPO's initial (neutron source to sample) 9-m-long flight path (Fig. 1). A T_o chopper removes the fast neutrons, or neutrons of a particular wavelength,



▲ *Fig. 1. Exploded view of HIPPO showing sample chamber surrounded by five conical three-dimensional rings of ^3He detector tubes in 10 atm. A white beam made up of pulsed neutrons of different energies (entering from the left) travels down a collimator to a chopper that cuts out very fast neutrons, allowing only slower thermal neutrons to continue down the flight path to the bulk material contained inside a 29.13-in.-diam sample chamber. The neutrons interact with the lattice (crystal) structure of the bulk material, diffract off, and impinge on the detectors. We measure the neutron diffraction (in the form of light signals) to ascertain how the energies or momentum of the neutrons changed after interacting with the atoms*

produced at the neutron-production target, and a collimator guides the beam down the length of the flight path toward the sample chamber. Scappers—plates consisting of multiple holes varying in size from large to small (from about 8 in. down to 2 in. in diameter)—gradually reduce the size of the beam as it travels toward the sample chamber and make it more coherent and focused. As the beam penetrates the bulk material in the sample chamber, neutrons interact with the atoms that make up the material and diffract off into the three-dimensional rings of detector banks, which are set at very specific angles. An array of about 1,400 ^3He detector tubes cover nearly a 4.6 m^2 area with five detector banks at scattering angles ranging from backscattering at nominally 150° to forward scattering at nominally 10°. The detector panels are tilted relative to the diffracted neutron paths to give a more constant resolution across their surfaces. The tilt compensates for the change in resolution caused by the angular placement of each panel with a corresponding change in the sample-to-detector flight path. The interaction between the neutrons and ^3He in the detector tubes produces ^4He plus gamma radiation and ionizes the gas, creating a cascade of electrons with associated charges. These charges are digitized and converted electronically to data and patterns of intensity versus scattering angle.

HIPPO's extremely high count rate makes it ideal for studying time-dependent processes and small and large samples of material under a wide variety of environmental conditions. Its three-dimensional arrangement of detectors will allow researchers to make direct measurements of the orientation of crystals in polycrystalline samples either without rotating the sample or using only very few sample rotations. An innovative "sample changer" will hold 110 samples at a time for rapid powder-diffraction measurements and 64 samples at a time for rapid texture measurements (Fig. 2). A complete measurement of the crystal orientation distribution in a typical texture experiment of, for example, a 1-cm stainless steel cube will take only about five minutes to make. This measurement time will vary depending on the size, weight, and composition of the sample. A number of environmental stages, including a cryostat, furnace, a three-axis texture goniometer with Kappa geometry, high-pressure cells, and sample changer, can be accommodated within HIPPO. Data acquisition will be based on current VME (virtual memory extension) technology and will make use of Web-based visualization and control software. Experiments can be controlled remotely from the user's laboratory.



▲ *Fig. 2. Kristin Bennett, HIPPO project manager, loads a texture sample on the goniometer in the multi-position sample changer for diffraction measurements.*

Myriad Research Applications with HIPPO

We designed HIPPO to accommodate various research efforts in obtaining *in situ* neutron-diffraction data. HIPPO can be applied to research in kinetics of reactions; high-pressure investigations of complex systems with large sample volumes; the evolution of texture in polycrystals during deformation processes; and recrystallization and phase transformation studies. TOF neutron-diffraction techniques offer some unique advantages over other methods in characterizing texture. Diffraction signals average over large volumes rather than surfaces, making grain statistics for texture analysis easily obtainable. Also, because intensity corrections are generally unnecessary with this method, a high level of accuracy can be obtained.[3] And the low-absorption property of neutrons allows researchers to use environmental stages (high temperature, low temperature, and bulk stress-strain) to observe *in situ* texture changes.[4,5] For pulsed polychromatic neutrons, a whole range of wavelengths is available, and continuous spectra can be recorded at moderate resolution. These types of neutrons are especially adaptive to problems of low crystal symmetry and composites with many diffraction peaks typical for modern materials and for many environmental problems.[6] The texture of such materials cannot be satisfactorily characterized by x-ray diffraction. New crystallographic methods are being used to analyze these spectra and obtain simultaneous information on crystal structure, orientation distribution, and phase proportions (from intensities), internal elastic strains (from peak locations), and microstructure (from peak shapes).[1, 2, 7] Areas of research might include texture and anisotropy studies of rocks, for example, granite-mylonite and mantle peridotites; crystal-structure studies of zeolites; and structure studies of liquids and melts, including aluminum-silicon melts and glasses.

In situ high proton-tritium (P-T) neutron-diffraction experiments provide a unique opportunity to study texture, hydrogen bonding, magnetic moments, and structural and thermal parameters of light elements (for example, hydrogen, lithium, and carbon) and heavy elements (for example, tantalum, uranium, and plutonium), which are virtually impossible to determine by x-ray diffraction techniques. For example, we can derive thermoelasticities and Debye-Waller factors as functions of pressure and temperature using *in situ* high P-T neutron-diffraction techniques. These applications can also be extended to a much broader spectrum of scientific problems. For instance, puzzles in Earth science, such as the carbon cycle and the role of hydrous minerals for water exchange between lithosphere and biosphere, can be directly addressed. Moreover, by introducing *in situ* shear strain, texture accompanied with phase transitions in the deep mantle can also be studied by high P-T neutron-diffraction experiments. With the new HIPPO toroidal anvil press (TAP-98) designed by Yusheng Zhao (LANSCE-12),[8] we will be able to accommodate *in situ* P-T neutron-diffraction experiments up to 30 GPa and 2000 K simultaneously. Sample sizes may range from 50 to 300 mm^3. The TAP-98 is positioned in the HIPPO sample chamber at different diffraction settings. A vertical setting will serve the purpose of studying texture development under high-pressure and high-temperature conditions. The TAP-98 will also provide a transmitting port for small-angle neutron-diffraction experiments.

Neutron scattering is the prime tool when it comes to determining spatial and temporal distributions of magnetization at the microscopic scale.[9] Magnetic scattering is due to a dipole interaction between the magnetic moments of the neutron and the unpaired outer-shell electrons, and it gives rise to additional contributions in the diffraction pattern. Therefore, neutrons offer the unique capability to measure spin, charge, and lattice effects simultaneously. Studies of magnetic scattering are effectively restricted to d-spacings greater than about 1 to 2 Å, and the HIPPO diffractometer covers significantly larger d-spacings. In addition, the large neutron flux of HIPPO will allow relatively fast measurements of magnetic materials, which often suffer from the fact that the magnetic intensities are weak compared to the nuclear ones. Finally, the HIPPO diffractometer provides novel capabilities, for example, the study of magnetic systems under pressure.

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